

Radionuclides in the Lichen–Caribou–Human Food Chain Near Uranium Mining Operations in Northern Saskatchewan, Canada

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The richest uranium ore bodies ever discovered (Cigar Lake and McArthur River) are presently under development in northeastern Saskatchewan. This subarctic region is also home to several operating uranium mines and aboriginal communities, partly dependent upon caribou for subsistence. Because of concerns over mining impacts and the efficient transfer of airborne radionuclides through the lichen–caribou–human food chain, radionuclides were analyzed in tissues from 18 barren-ground caribou (*Rangifer tarandus groenlandicus*). Radionuclides included uranium (U), radium (²²⁶Ra), lead (²¹⁰Pb), and polonium (²¹⁰Po) from the uranium decay series; the fission product (¹³⁷Cs) from fallout; and naturally occurring potassium (⁴⁰K). Natural background radiation doses average 2–4 mSv/year from cosmic rays, external gamma rays, radon inhalation, and ingestion of food items. The ingestion of ²¹⁰Po and ¹³⁷Cs when caribou are consumed adds to these background doses. The dose increment was 0.85 mSv/year for adults who consumed 100 g of caribou meat per day and up to 1.7 mSv/year if one liver and 10 kidneys per year were also consumed. We discuss the cancer risk from these doses. Concentration ratios (CRs), relating caribou tissues to lichens or rumen (stomach) contents, were calculated to estimate food chain transfer. The CRs for caribou muscle ranged from 1 to 16% for U, 6 to 25% for ²²⁶Ra, 1 to 2% for ²¹⁰Pb, 6 to 26% for ²¹⁰Po, 260 to 370% for ¹³⁷Cs, and 76 to 130% for ⁴⁰K, with ¹³⁷Cs bio-magnifying by a factor of 3–4. These CRs are useful in predicting caribou meat concentrations from the lichens, measured in monitoring programs, for the future evaluation of uranium mining impacts on this critical food chain. **Key words:** caribou, cesium, food chain, lead, polonium, radiation dose, radionuclides, radium, risk, uranium. *Environ Health Perspect* 107:527–537 (1999). [Online 27 May 1999]

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The most critical food chain in the world for concentrating airborne radionuclides is the lichen–caribou–human food chain. Lichens accumulate atmospheric radionuclides more efficiently than other vegetation due to their lack of roots, large surface area, and longevity. Uptake from the substrate is minimal compared with the uptake from wet or dry deposition (1,2). Lichens are the main winter forage for caribou, which in turn, are a main dietary staple for many northern Canadians. Thus, airborne radionuclides, particularly cesium-137 (¹³⁷Cs), lead-210 (²¹⁰Pb), and polonium-210 (²¹⁰Po), are transferred efficiently through this simple food chain to people, elevating their radiological dose (3–6).

Of these three radionuclides, ²¹⁰Po delivers the greatest dose, contributing 57–72% of the background radiation dose to aboriginal residents in the Canadian Arctic, primarily from the consumption of caribou meat. Annual doses ranged from 2 to 14 mSv/year in adults and children, which was determined using the dose conversion factors (DCFs) 8.8×10^{-6} Sv/Bq for children and 1.2×10^{-6} Sv/Bq for adults (7) and assuming ingestion rates of 29–568 kg/year in adults and 7.5–93.7 kg/year in children (8).

Generally, concentrations of ²¹⁰Po in caribou meat increase as one moves north

and east across the Canadian Arctic. The Beverly herd of central Canada has ²¹⁰Po concentrations of 15 Bq/kg wet weight in their winter range near Snowdrift and 17 Bq/kg near Baker Lake (3,4). Concentrations as high as 40 Bq/kg have been reported further to the northeast (9).

The Beverly herd of central Canada occasionally enters the subarctic boreal forest of northeastern Saskatchewan, where the caribou may winter near a number of operating and proposed uranium mines. Two new mines, Cigar Lake and McArthur River, south and west of Wollaston Lake (Figure 1), are currently being developed and have the richest ore grades in the world.

The mining and milling of uranium ores release radionuclides from the uranium-238 series (Table 1) to the terrestrial environment via crushing and grinding of ore, wind erosion of tailings, and emanation of radon gas. The most persistent radionuclides have the longest half-lives; thus, U in ore dusts, ²²⁶Ra and ²¹⁰Pb in tailings dusts, and ²¹⁰Pb and ²¹⁰Po aerosols from radon gas decay are of greatest concern. The potential for enhanced atmospheric deposition of these radionuclides onto lichens in the mining area could elevate radiation doses in both caribou and in people consuming these caribou. Thus, this

study was undertaken to establish radionuclide concentrations in caribou before any further mining developments proceed.

Barren-ground caribou (*Rangifer tarandus groenlandicus*) from the Beverly herd entered the Wollaston Lake area of Saskatchewan in January–March 1995 (Figure 1). The last time caribou wintered in the area was in 1980–1981. In most years, local residents must travel north near or into the Northwest Territories to hunt caribou. The presence of caribou presented a unique opportunity to measure uranium and its decay products while the animals were on winter range, relatively close to uranium mines and the aboriginal community of Wollaston Post.

Materials and Methods

On 14–15 March 1995, 18 caribou (15 females and 3 males from 2 to 12 years of age) were collected approximately 60 miles north of the community of Wollaston Post in the Charcoal Lake/ Cochrane River area (latitude 59°08–12'N, longitude 102°10–13'W) by local community hunters. The local hunters, as treaty Indians, traditionally harvest caribou without quota or any hunting season restrictions. In this study, the meat from the 18 caribou was donated to the Wollaston Post community at large, and provincial government personnel and veterinarians were present to aid with dissection of remaining tissues.

The tissue samples were collected in the field, including bone, liver, kidney, muscle, fur, feces, blood, spleen, lung, pancreas, and components of the gastrointestinal (GI) tract; the samples were shipped to the University of Saskatchewan in Saskatoon and prepared for radiochemical analyses. Rings of cortical bone samples were sawed from the midfemur after removal of marrow

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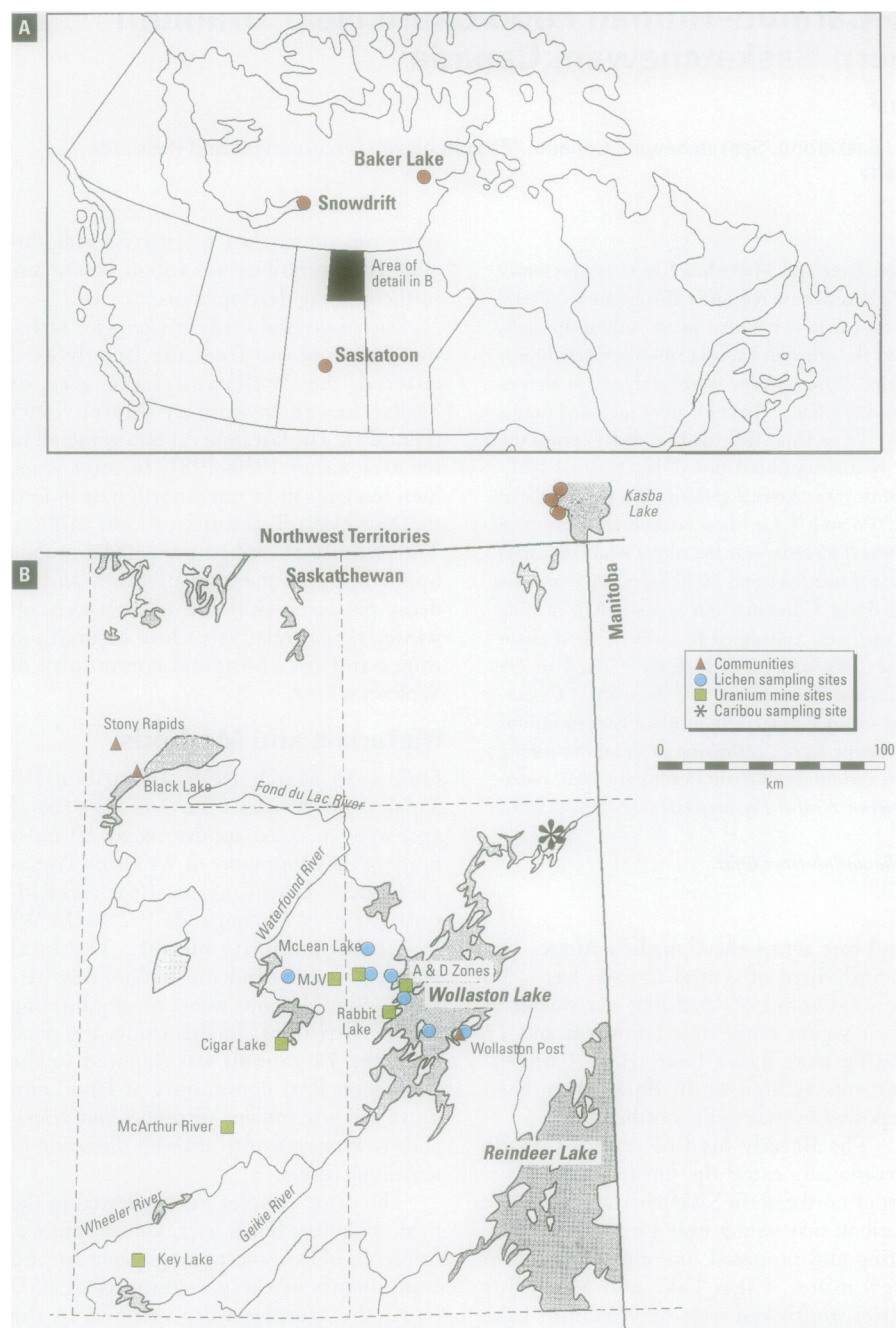


Figure 1. Maps of (A) Canada and (B) the northeastern Saskatchewan study area, including caribou and lichen sampling sites and uranium mine sites.

and excess tissue. Kidney samples consisted of both cortex and medulla. All muscle samples were from the gastrocnemius. The GI tract wall samples were rinsed with deionized water to remove contents. For most tissue types, one or two blind duplicates were prepared, representing 8–25% of the samples available for each tissue type and radionuclide.

Radiochemical analyses were performed at the Saskatchewan Research Council (SRC) in Saskatoon, a commercial laboratory that analyzes the majority of samples

from Saskatchewan mining operations and environmental monitoring programs. Following gamma spectroscopy, most samples were digested in nitric acid and split into three portions for *a*) ^{210}Po and ^{210}Pb analyses; *b*) ^{226}Ra analyses; and *c*) U analyses. A portion of the samples was desiccated prior to digestion to obtain constant dry weights. Liver and muscle samples were dry ashed at 500° and the ash was split for U, ^{226}Ra , and ^{210}Pb , with the portion for ^{210}Po analyses requiring separate digestion in nitric acid.

Table 1. Uranium-238 decay series.

Radionuclide	Principle radiation type	Half-life
Uranium-238	Alpha	4.5 billion years
Thorium-234	Beta	24.1 days
Protactinium-234	Beta	114 min
Uranium-234	Alpha	235,000 years
Thorium-230	Alpha	80,000 years
Radium-226	Alpha	1,620 years
Radon-222 gas	Alpha	3.85 days
Polonium-218	Alpha	3.05 min
Lead-214	Beta	26.8 min
Bismuth-214	Beta	19.7 min
Polonium-214	Alpha	0.00015 sec
Lead-210	Beta	22.2 years
Bismuth-210	Beta	4.97 days
Polonium-210	Alpha	138 days
Lead-206	—	Stable

Analytical procedures followed methods described by the Canadian Centre for Mineral and Energy Technology (10). Uranium was determined by either kinetic phosphorescence analysis (KPA) [detection limit (DL) = 0.1 ppb or 0.1 ng/g] or by mass spectroscopy (DL = 1 ppb) for cases where samples yielded colored solutions after digestion. DLs were defined as those concentrations where error is 100%, as a function of background counts, tracer recovery, and counting efficiency. DLs are calculated as follows:

DL for ^{210}Po or ^{226}Ra =

$$\frac{3 \times \text{background counts}^{1/2}}{\text{counting time in minutes}} \times \frac{1 \text{ min}}{60 \text{ sec}} \times \frac{\text{counts for 100\% recovery of tracer}}{\text{tracer counts} - \text{background counts}} \times \frac{1}{\text{sample weight (grams)}} \times \frac{1}{\text{counting efficiency}}$$

Counting efficiencies are 0.343 for ^{210}Po by alpha spectroscopy, but 0.22 for ^{226}Ra precipitated on disk, which must be placed further from the detector. Uranium DLs by delayed neutron counting (DNC) are based on terms for background counts, an ash weight/wet weight conversion factor, and a calibration factor of 102 counts per minute (cpm) per microgram of uranium. Detection limits for ^{210}Pb include terms for *a*) background counts of ^{210}Bi ; *b*) 40 ppm stable Bi tracer for 100% recovery/measured ppm in sample; *c*) $1/e^{-\lambda t}$, where $e^{-\lambda t}$ = the decay of ^{210}Bi over time t (in days) from extraction to midpoint of counting time and $\lambda = 0.693/\text{half-life of } ^{210}\text{Bi}$ (5.3 days); and *d*) a calibration factor

based on standards, i.e., $1/(\text{corrected cpm/Bq } ^{210}\text{Bi})$.

Uranium in dry-ashed samples of liver or muscle was determined by delayed neutron counting (DL = 100 ppb) using a Slowpoke II reactor at the SRC in Saskatoon. ^{226}Ra was analyzed as a coprecipitate of barium sulfate (DL = 0.005

Bq/g) using gross alpha counting after 6 days of ingrowth of the ^{226}Ra progeny.

^{210}Po was plated for 6 hr from 0.5M HCl solution onto silver disks and counted by alpha spectrometry for 100 min (DL = 0.005 Bq/g), using ^{208}Po as a tracer to estimate recovery. After ^{210}Po plating, the depleted ^{210}Po solution was analyzed for

^{210}Pb using a bismuth-oxychloride precipitation of the ^{210}Pb decay product, ^{210}Bi (DL = 20 Bq/kg). Detection limits were based on 20 g samples with samples > 20 g improving detectability.

For samples below detection limits, 50% of the detection limit was used to estimate the concentration. The ^{210}Po data

Table 2. Radionuclide concentrations (wet weight) in Wollaston caribou tissues.

Parameter	Unat ^a (ppb)	Unat (Bq/kg)	^{226}Ra (Bq/kg)	^{210}Pb (Bq/kg)	mPo (Bq/kg)	tcPo (Bq/kg)	U/Ra ratio	tcPo/Pb ratio	^{137}Cs (Bq/kg)	^{40}K (Bq/kg)	Cs/ ^{40}K ratio	dw as % of ww
Bone												
Geometric mean	23 ^b	0.57 ^b	69	651	352	169	0.008	0.26	58	18 ^b	3.1	100% assumed
Arithmetic mean	26 ^b	0.67 ^b	72	669	367	217	0.011	0.331	59	27 ^b	4.2	
SE	4	0.11	5	37	25	27	0.003	0.037	4	8	0.8	
n	18 (14)	18 (14)	18	18	18	18	18	18	11	11 (7)	11	
Liver												
Geometric mean	1.9	0.048	0.81	138	275	316	0.057	2.28	228	80	2.8	
Arithmetic mean	2.3	0.057	1.7	154	286	332	0.13	2.59	232	81	3.0	30%
SE	0.3	0.008	0.6	18	19	25	0.04	0.30	13	5	0.3	5%
n	18	18	11	18	18	18	11	18	11	11	11	11
Kidney												
Geometric mean	14 ^b	0.35 ^b	1.1	155	154	142	0.49	0.91	553	85	6.5	
Arithmetic mean	18 ^b	0.44 ^b	1.2	169	159	156	0.60	1.26	557	89	6.9	20%
SE	4	0.10	0.2	16	11	18	0.21	0.27	20	8	0.8	0.4%
n	18 (15)	18 (15)	5	18	18	18	5	18	11	11	11	5
Muscle												
Geometric mean	0.69 ^b	0.02 ^b	0.16	1	12	14	0.61	22	367	76	4.9	
Arithmetic mean	1.6 ^b	0.040 ^b	0.21	1.1	12.4	14.0	1.6	35	370	78	4.9	27%
SE	0.6	0.014	0.07	0.3	0.5	0.6	1.2	9	12	4	0.2	1%
n	18 (13)	18 (13)	4	18	18	18	4	18	18	18	18	4
Weighted average												
Geometric mean	2.3	0.057	3.7	36		27	0.015	0.750	345	72	4.7	
Arithmetic mean	2.9	0.072	3.8	37		28	0.022	0.783	347	74	4.7	
SE	0.5	0.014	0.2	2		2	0.005	0.055	11	4	0.2	
n	18	18	18	18		18	18	18	18	18	18	
Rumen contents												
Geometric mean	10 ^b	0.25 ^b	0.82	81	125	148	0.31	1.8	99	66	1.5	
Arithmetic mean	10 ^b	0.25 ^b	0.83	83	128	153	0.31	1.9	101	73	1.6	18%
SE	NA	NA	0.04	4	6	10	0.01	0.15	6	7	0.2	0.5%
n	18 (18)	18 (18)	12	18	18	18	13	18	18	18	18	18
Feces												
Geometric mean	77	1.9	3.0	452	335	304	0.28	0.67	230	26 ^b	8.9	
Arithmetic mean	97	2.4	3.1	469	368	344	0.30	0.71	231	40 ^b	13	36%
SE	15	0.4	0.4	33	47	53	0.05	0.06	12	19	6	1%
n	16	16	5	17	17	17	5	17	4	4 (2)	4	17
Blood												
Geometric mean	4.5 ^b	0.11 ^b	0.61	11	22	26	0.22	2.3	41	37	1.1	
Arithmetic mean	4.8 ^b	0.12 ^b	0.64	14	23	28	0.23	2.5	42	44	1.3	23%
SE	NA	NA	0.09	2	2	3	0.04	0.3	2	5	0.2	1%
n	10 (10)	10 (10)	5	17	17	17	5	17	17	17 (1)	17	5
Fur												
Geometric mean	11 ^b	0.28 ^b	0.37 ^b	400	55	63	0.69	0.16	79	37	2.1	
Arithmetic mean	12 ^b	0.30 ^b	0.40 ^b	465	58	65	0.80	0.18	84	38	2.3	52%
SE	NA	NA	0.08	78	5	6	0.20	0.03	15	4	0.4	2%
n	5 (5)	5 (5)	5 (3)	11	18	11	5	11	5	5	5	18
Spleen mean	29 ^b	0.72 ^b		7.0	30	35		5.1				22%
SE	15	0.37		0.8	3.9	4.2		0.4				0%
n	2 (1)	2 (1)		6	6	6		6				2
Rumen wall mean			11.1	32	36		3.4					13%
SE			1.8	5.5	5.7		0.5					2%
n			6	6	6		6					2
Duodenum mean	8.0	0.20		19.4	42	49		2.7	170	35	9.9	
SE	5.0	0.1		2.8	4.2	6.2		0.5	0	14	4.1	
n	2	2		6	6	6		6	6	6	6	
Colon mean (n = 2)			3.2	11.0	12.2		7.5					
Lung (n = 1)	17	0.43		10	25	31		3.15				19%
Pancreas (n = 1)			15	60	79		5.29					13%

Abbreviations: DL, detection limit; dw, dry weight; mPo, measured ^{210}Po ; NA, not applicable; SE, standard error; tcPo, time-corrected ^{210}Po ; Unat, natural uranium; ww, wet weight. Values in parentheses are the number of samples below DL; 50% of the DL was assigned to estimate the value in mean calculations.

^aUnat is composed of 0.711% ^{235}U , 99.2837% ^{238}U , and 0.0053% ^{234}U by weight and is converted to Bq/kg by multiplying by 0.0252. ^bTissues for which $\geq 50\%$ of the analyses were below DLs.

were time corrected to account for decay of unsupported ^{210}Po and ingrowth from ^{210}Pb between the date of kill and the date of alpha spectroscopy, as follows:

$$[\text{tcPo}] = [\text{mPo}] - [\lambda\text{Po}/(\lambda\text{Po} - \lambda\text{Pb})] \\ \times [\text{mPb} \times (e^{\lambda\text{Pb} \times t} - e^{\lambda\text{Po} \times t})] \\ \times 1/e^{\lambda\text{Po} \times t},$$

where $\text{tcPo} = ^{210}\text{Po}$ concentrations, time-corrected from the date of sampling to the date of ^{210}Po analysis; $\text{mPo} =$ measured ^{210}Po concentrations at time t ($t =$ time in days between date of kill and date of ^{210}Po analysis); mPb is ^{210}Pb concentrations measured at time t ; $\lambda =$ the radiological decay constant or fraction of the initial amount of the radionuclide decaying per day (e.g., λPo for $^{210}\text{Po} = 0.693/138$ days $= 0.0050217$ days $^{-1}$, and λPb for $^{210}\text{Pb} = 0.693/(22.2 \text{ years} \times 365.25 \text{ days/year}) = 0.000855$ days $^{-1}$). These time corrections have proven valuable as a means to check laboratory accuracy because negative ^{210}Po concentrations after time corrections indicate cases in which ^{210}Pb measurements are erroneously high or measured ^{210}Po values are erroneously low.

Food chain transfer was estimated by concentration ratios (CRs). A weighted average (WA) of caribou tissues was calculated from bone, liver, kidney, and muscle concentrations, where $\text{WA} = 0.0597 [\text{bone}] + 0.0106 [\text{liver}] + 0.0022 [\text{kidney}] + 0.9275 [\text{muscle}]$. This equation was based on percentage of total body weight (11). Weighted average concentrations, as well as muscle concentrations, were divided by concentrations in rumen (stomach) contents or in lichens, used as an estimate of the caribou food source, to obtain CR values.

The CRs, using rumen contents, were determined for individual caribou on both a

wet and a dry weight basis. Dry weight concentrations were obtained by dividing wet weight concentrations by an empirically determined dry/wet ratio for each tissue type.

Two sets of lichen data were used to estimate the food chain transfer to the 1995 Wollaston caribou: *a*) 12 samples of the most abundant caribou lichen, *Cladina mitis*, sampled in 1991 at Kasba Lake north of where the caribou were shot and *b*) 8 samples of *Cladina stellaris*, collected close to the Wollaston mining area in 1994 (12).

Tissue concentrations and food chain CRs for ^{210}Pb and ^{210}Po for Wollaston caribou were compared to previous data from Baker Lake and Snowdrift (now called Lutsel K'e) in the Northwest Territories (3,4). One-way analysis of variance (ANOVA) tests were performed on both arithmetic and \log_{10} -transformed data. Where assumptions of normality and equal variance failed, Kruskal-Wallis ANOVAs were performed on ranked data. Significant differences between regional means were determined by Tukey's multiple range tests. All statistical tests were performed using SigmaStat2 computer software (13).

Results

Concentrations of uranium series radionuclides. Mean radionuclide concentrations (wet weight) in caribou tissues are summarized in Table 2. Uranium was detectable in feces (97 ppb), blood (4.8 ppb), and liver (2.3 ppb) (Figure 2). All samples of rumen (stomach) contents and fur and most samples of bone, kidney, and muscle were below detection limits, so 50% of detection limits were assumed for calculations. Uranium in parts per billion was converted to becquerels per kilogram by multiplying by 0.0252 to account for the

activities of ^{235}U , ^{234}U , and ^{238}U in naturally occurring uranium.

^{226}Ra was highest in bone (72 Bq/kg), ranged from 0.23 to 1.7 Bq/kg in other tissues, and was up to 3.1 Bq/kg in feces (Figure 3). The even distribution of ^{226}Ra in caribou tissues other than bone was similar in magnitude to concentrations in Saskatchewan prairie rodents (14), indicating that these ^{226}Ra concentrations were not particularly elevated in caribou from the Wollaston region.

^{210}Pb concentrations were > 400 Bq/kg in bone, fur, and feces and as low as 1 Bq/kg in muscle. Figure 4 compares bone, liver, kidney, muscle, weighted average and rumen content concentrations in Wollaston caribou tissues versus concentrations in the same tissues previously collected from Baker Lake and Snowdrift caribou (Figure 1) (3,4). One-way ANOVA and multiple range tests showed that caribou from the tundra region near Baker Lake had significantly higher ^{210}Pb concentrations in bone and rumen contents than caribou from the forested regions near Snowdrift or Wollaston ($p < 0.001$). However, Wollaston caribou kidneys contained significantly more ^{210}Pb than either Baker Lake or Snowdrift caribou ($p < 0.001$). The high ^{210}Pb in kidneys may reflect short-term increases in ^{210}Pb intake for caribou in the Wollaston mining area, but these increases would not affect long-term storage of ^{210}Pb in bone. The high levels of ^{210}Pb in Wollaston caribou fur relative to soft tissues may reflect surface adsorption of aerially deposited ^{210}Pb because fur has a large surface area for atmospheric deposition, just as large surface areas elevate ^{210}Pb levels in lichens.

Age in caribou showed little correlation with concentrations of the bone-seeking

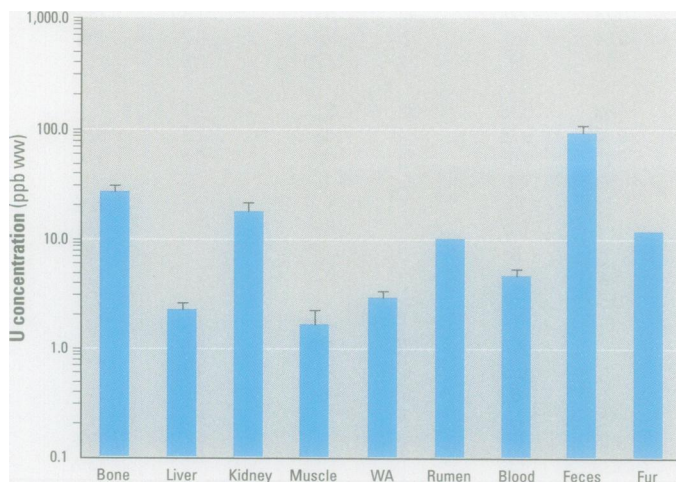


Figure 2. Uranium concentrations (mean and standard error) in Wollaston caribou tissues. Abbreviations: WA, weighted average; ww, wet weight.

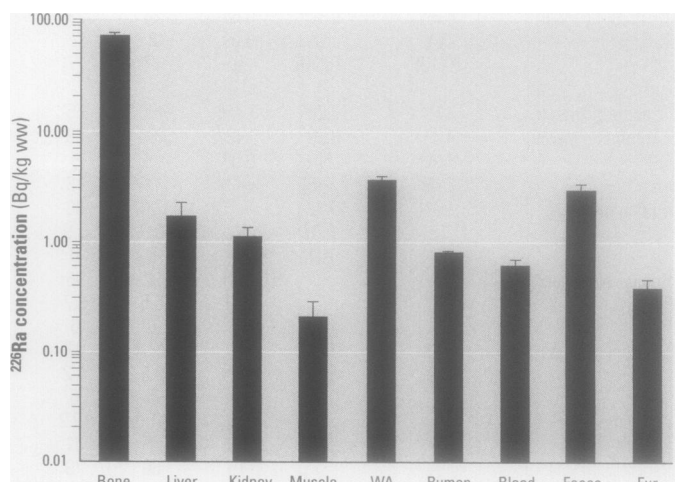


Figure 3. ^{226}Ra concentrations (mean and standard error) in Wollaston caribou tissues. Abbreviations: WA, weighted average; ww, wet weight.

radionuclides ^{226}Ra and ^{210}Pb in bone. Linear regression procedures (Model I) yielded the following equations and insignificant r^2 values: Bone = $-1.73 \text{ age} + 81$ for ^{226}Ra ($r^2 = 0.0602$, $p = 0.32$) and Bone = $-3.5 \text{ age} + 684$ for ^{210}Pb ($r^2 = 0.0028$, $p = 0.83$) by Model I regression techniques. The loss of these radionuclides in caribou bone over time is attributed to bone turnover as well as the growth and drop of antlers every year.

Polonium-210 was $> 300 \text{ Bq/kg}$ in bone, liver, and feces, approximately 150 Bq/kg in kidney and rumen contents, and an order of magnitude lower in muscle and blood (Figure 5). Wollaston caribou generally had concentrations lower than Baker Lake caribou but higher than Snowdrift caribou. Significant differences ($p < 0.001$) in ^{210}Po concentrations, according to ANOVA on arithmetic, log-transformed, and ranked data, included *a*) higher levels in bone and kidney in Baker Lake versus Snowdrift caribou; *b*) higher levels in Baker Lake and Wollaston caribou liver versus Snowdrift; and *c*) higher weighted average concentrations in Baker Lake animals versus both Wollaston and Snowdrift animals.

Isotopic ratios. Isotopic ratios were also calculated to see how radionuclide distribution and thus internal radiation doses differ among tissues (Figure 6). With the exception of low ratios in bone, $\text{U}/^{226}\text{Ra}$ ranged from 0.22 in blood to 0.49 in kidney to 0.69 in fur (Table 1), indicating the affinity of ^{226}Ra for bone and the relatively high accumulation of U in kidney. Liver, muscle, blood, and limited data for spleen, lung, pancreas, and intestinal wall tissues had $^{210}\text{Po}/^{210}\text{Pb}$ ratios above unity, indicating the affinity of ^{210}Po for soft tissues beyond amounts that could arise from the decay of ^{210}Pb within that tissue. When mean $^{210}\text{Po}/^{210}\text{Pb}$ ratios are compared in caribou tissues from Baker Lake, Snowdrift, and Wollaston Lake, Wollaston kidney and potentially bone exhibit low ratios (Table 3).

The low $^{210}\text{Po}/^{210}\text{Pb}$ ratio found for Wollaston caribou kidneys reflects the significantly higher ^{210}Pb levels found, relative to Baker Lake and Snowdrift caribou kidneys.

All three regions show $^{210}\text{Po}/^{210}\text{Pb}$ ratios of approximately 2 in rumen contents, in contrast to lichens, the winter food source for caribou, which have ratios < 1 . The high ratio in rumen contents suggests that caribou either secrete ^{210}Po into rumen contents with saliva and/or that more ^{210}Pb than ^{210}Po is absorbed from the rumen during digestion. Further down the digestive tract, $^{210}\text{Po}/^{210}\text{Pb}$ ratios dropped to 1.0 in the lower stomach compartment (abomasum), rose again in the duodenum to 2.7,

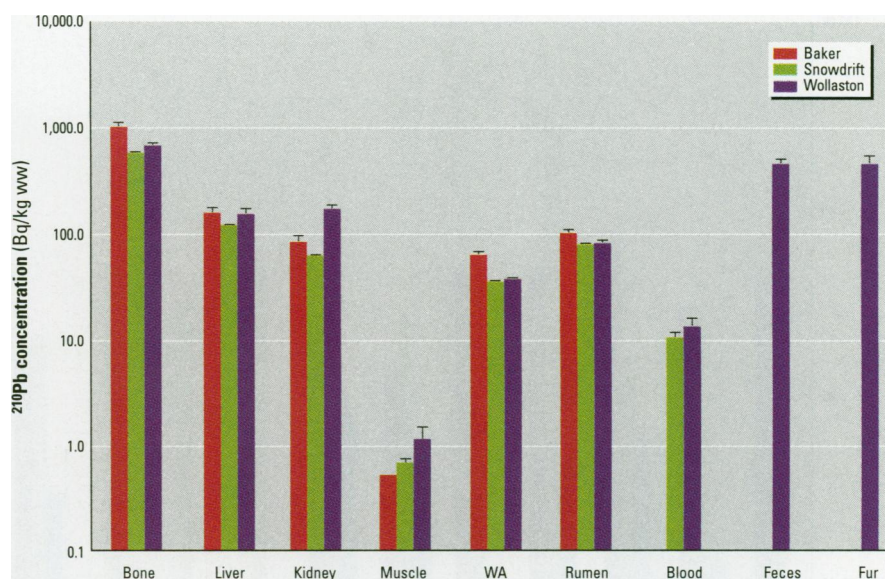


Figure 4. ^{210}Pb concentrations (mean and standard error) in Baker Lake ($n = 24$), Snowdrift ($n = 23$), and Wollaston ($n = 18$) caribou tissues. Abbreviations: WA, weighted average; ww, wet weight.

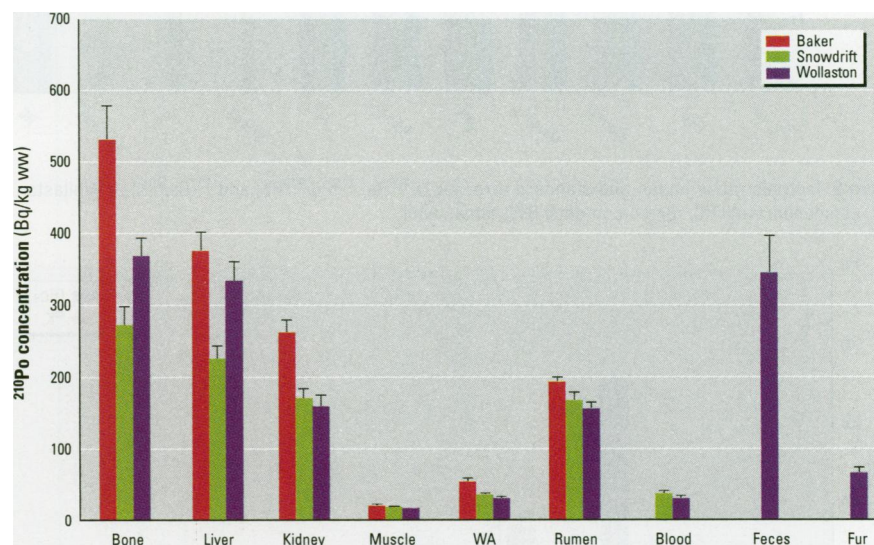


Figure 5. ^{210}Po concentrations (mean and standard error) in caribou tissues from Baker Lake ($n = 24$), Snowdrift ($n = 23$), and Wollaston ($n = 18$). Abbreviations: WA, weighted average; ww, wet weight.

and then fell in the feces to 0.7. These ratios and those in the GI tract wall of the rumen (3.4), abomasum (2.9), and rectum (7.5) suggest that more ^{210}Pb is absorbed in the upper GI tract and more ^{210}Po in the lower GI tract.

Concentrations of ^{137}Cs , ^{40}K , and other gamma-emitting radionuclides. Gamma spectroscopy was performed on the Wollaston caribou tissues to determine concentrations of the fission product ^{137}Cs . Cesium has been an important contributor to radiological doses in the lichen–caribou–human food chain due to atmospheric fallout from nuclear weapons tests in the 1960s and from the 1986 Chernobyl nuclear accident. Naturally occurring

potassium-40 (0.01% of all K) was also measured because the availability of K affects the uptake and distribution of ^{137}Cs , a K analog. Concentrations of ^{137}Cs were highest in kidney (557 Bq/kg) and muscle (370 Bq/kg) and lowest in blood (42 Bq/kg), whereas ^{40}K ranged from 89 Bq/kg in kidney to 31 Bq/kg in bone (Figure 7). Mean $^{137}\text{Cs}/^{40}\text{K}$ ratios ranged from 1–2 in rumen contents and blood, to 4.7 in average caribou tissues (WA) to 9–13 in feces (Table 2, Figure 6). Gamma spectroscopy also yielded low but detectable amounts of some naturally occurring thorium-232 (^{232}Th) decay series radionuclides in 5 of the 11 caribou bones measured. The bone samples ranged from $< 10 \text{ Bq/kg}$ to 30 Bq/kg for

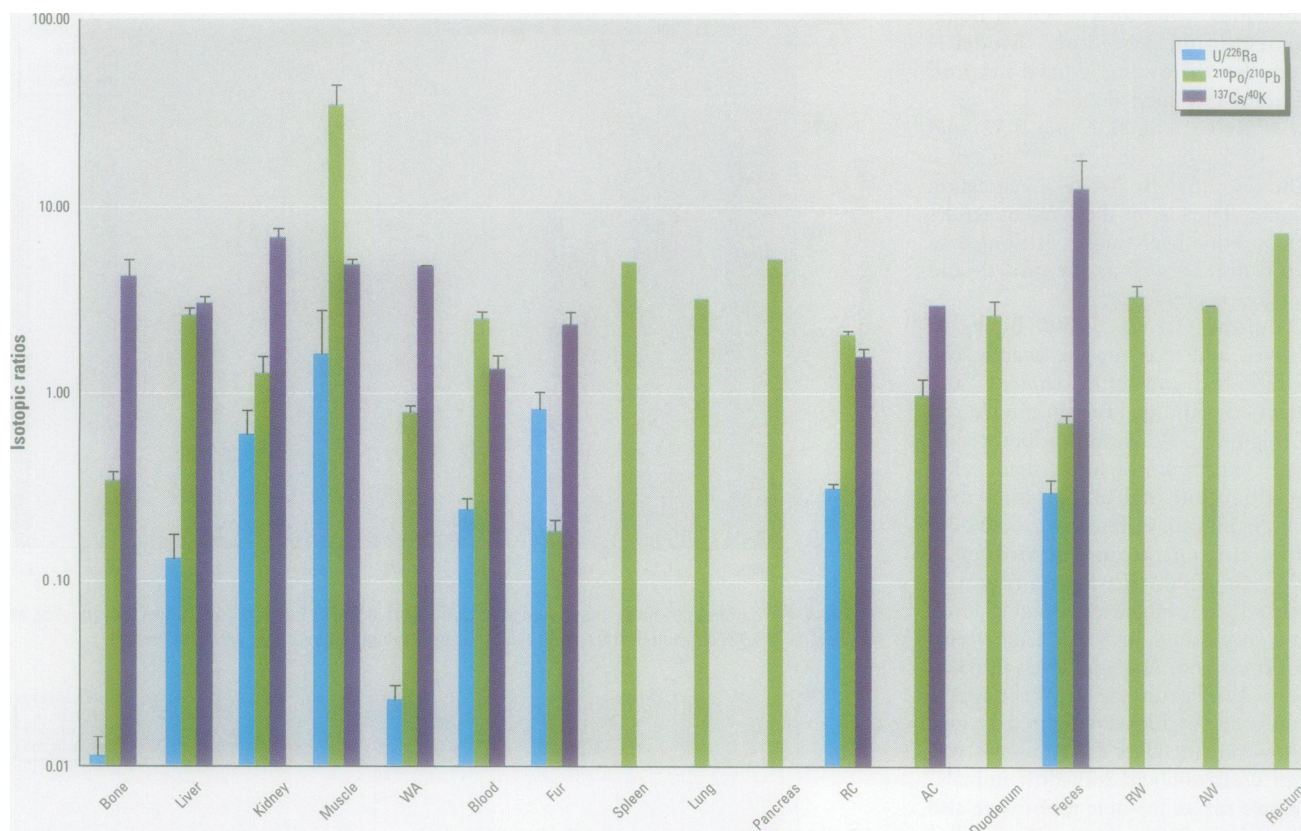


Figure 6. Isotopic ratios (mean and standard error for $U/^{226}Ra$, $^{210}Po/^{210}Pb$, and $^{137}Cs/^{40}K$) in Wollaston caribou tissues. Abbreviations: AC, abomasum contents; AW, abomasum wall; RC, rumen contents; RW, rumen wall.

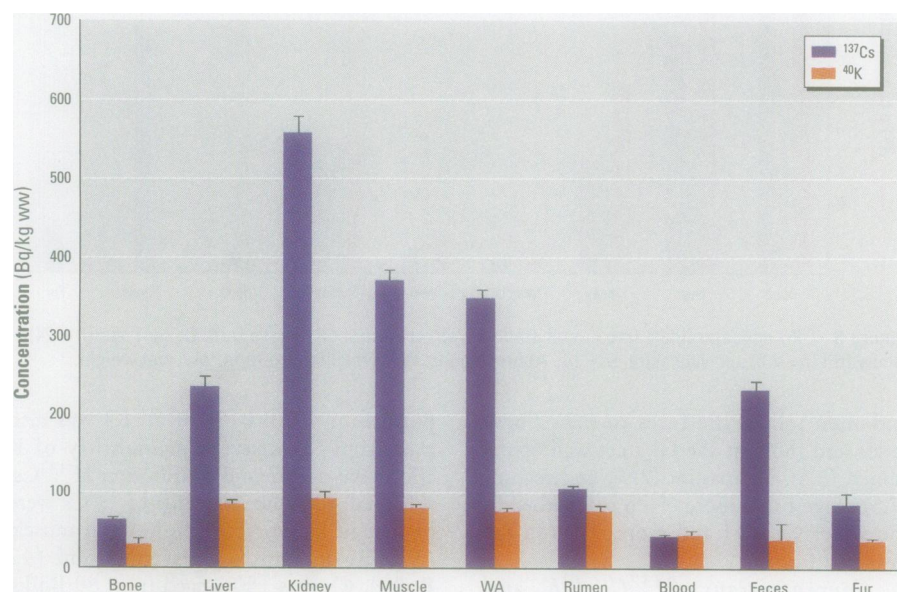


Figure 7. ^{137}Cs and ^{40}K concentrations (mean and standard error) in Wollaston caribou tissues. Abbreviations: WA, weighted average; ww, wet weight.

^{228}Ac , < 10 to 10 Bq/kg for ^{220}Rn and ^{212}Pb , and < 10 to 20 Bq/kg for ^{212}Bi .

Food chain transfer. CRs of average caribou tissue to rumen contents for each of the 18 Wollaston caribou were calculated on both a wet weight and a dry weight basis

(Table 4, Figure 8). The transfer rates showed that 1 kg of average caribou tissue (a WA of bone, liver, kidney, and muscle) contained 15–29% of the U, 320–510% of the ^{226}Ra , 30–45% of the ^{210}Pb , 12–19% of the ^{210}Po , 240–360% of the ^{137}Cs , and

Table 3. $^{210}Po/^{210}Pb$ ratios in caribou tissues from Baker Lake, Snowdrift, and Wollaston Lake.

Tissue	Baker Lake	Snowdrift	Wollaston
Bone	0.57	0.51	0.33
Liver	2.2	2.6	2.6
Kidney	3.6	3.3	1.3
Muscle	34	30	35
Rumen contents	2.0	2.3	1.9

73–130% of the ^{40}K found in 1 kg of dry rumen contents.

In every case, geometric means yielded the lowest CRs and arithmetic means when dry weight concentrations were used and yielded the highest CRs when wet weight concentrations were used. Wet weight CRs were higher than dry weight CRs because: *a*) wet weight rumen contents were “diluted” by saliva (only 17% dry weight), relative to caribou soft tissues, where muscle = 27% dry weight; and *b*) bone concentrations of ^{226}Ra and ^{210}Pb greatly elevated wet weight caribou concentrations relative to rumen contents.

The use of the lichen data from Table 5 lowered U and ^{226}Ra CRs but raised ^{210}Pb and ^{210}Po CRs relative to CRs based on rumen contents. If *Cladonia stellaris* lichens (sampled in 1994 around Wollaston Lake, west of where the caribou were collected)

Table 4. Food chain concentration ratios (CRs) for Wollaston caribou based on food source using either rumen contents or lichens.^a

Radionuclide	CR type	Weight type	Geometric mean	Arithmetic mean	Standard deviation	Standard error	n
Caribou WA Uranium	WA/rumen contents	ww	0.23	0.29	0.23	0.05	18
		dw	0.15	0.20	0.16	0.04	18
²²⁶ Ra	WA/ <i>C. stellaris</i>	dw	0.046	0.048			
		WA/rumen contents	ww	4.8	5.1	1.7	0.5
²¹⁰ Pb	WA/ <i>C. stellaris</i>	dw	3.2	3.4	1.1	0.3	12
		dw	1.5	1.3			
²¹⁰ Pb	WA/rumen contents	ww	0.44	0.45	0.11	0.03	18
		dw	0.30	0.31	0.07	0.02	18
²¹⁰ Po	WA/ <i>C. stellaris</i>	dw	0.42	0.42			
		WA/ <i>C. mitis</i>	dw	0.56	0.55		
²¹⁰ Po	WA/rumen contents	ww	0.18	0.20	0.08	0.02	18
		dw	0.12	0.13	0.06	0.01	18
¹³⁷ Cs	WA/ <i>C. stellaris</i>	dw	0.44	0.41			
		WA/ <i>C. mitis</i>	dw	0.51	0.49		
⁴⁰ K	WA/rumen contents	ww	3.5	3.6	1.0	0.24	18
		dw	2.4	2.5	0.8	0.20	18
Cs/ ⁴⁰ K	WA/rumen contents	ww	1.1	1.3	0.8	0.18	18
		dw	0.73	0.87	0.56	0.13	18
Cs/ ⁴⁰ K	WA/rumen contents	ww	3.1	3.4	1.4	0.33	18
		dw	2.1	2.3	1.1	0.25	18
Caribou muscle Uranium	Muscle/rumen contents	ww	0.069	0.16	0.24	0.06	18
		dw	0.046	0.11	0.16	0.04	18
²²⁶ Ra	Muscle/ <i>C. stellaris</i>	dw	0.014	0.027			
		Muscle/rumen contents	ww	0.18	0.25	0.17	0.09
²¹⁰ Pb	Muscle/ <i>C. stellaris</i>	dw	0.11	0.16	0.11	0.05	4
		dw	0.064	0.073			
²¹⁰ Pb	Muscle/rumen contents	ww	0.0079	0.013	0.014	0.003	18
		dw	0.0052	0.0086	0.0090	0.002	18
²¹⁰ Po (time-corrected)	Muscle/ <i>C. stellaris</i>	dw	0.0076	0.013			
		Muscle/ <i>C. mitis</i>	dw	0.010	0.017		
²¹⁰ Po (time-corrected)	Muscle/rumen contents	ww	0.093	0.098	0.029	0.007	18
		dw	0.062	0.064	0.019	0.004	18
¹³⁷ Cs	Muscle/ <i>C. stellaris</i>	dw	0.22	0.21			
		Muscle/ <i>C. mitis</i>	dw	0.26	0.25		
¹³⁷ Cs	Muscle/rumen contents	ww	3.7	3.9	1.1	0.3	18
		dw	2.5	2.6	0.9	0.2	18
⁴⁰ K	Muscle/rumen contents	ww	1.1	1.3	0.8	0.2	18
		dw	0.76	0.90	0.59	0.14	18
Cs/ ⁴⁰ K	Muscle/rumen contents	ww	3.2	3.6	1.5	0.4	18

Abbreviations: dw, dry weight; WA, weighted average (of bone, liver, kidney, and muscle); ww, wet weight.

^aLichen species include *Cladina stellaris* lichens from the Wollaston mining area and *C. mitis* from Kasba Lake, north of Wollaston Lake.

were used as the food source, then food chain CRs were 4.6–4.8% for U, 131–145% for ²²⁶Ra, 42–43% for ²¹⁰Pb, and 40–44% for ²¹⁰Po. If *Cladina mitis* lichens (sampled in 1991 from Kasba Lake, north of where the caribou were collected) were used, food chain CRs were 55–56% for ²¹⁰Pb and 49–51% for ²¹⁰Po. Geometric means yielded higher CRs than arithmetic means, which was not the case with the CRs based on rumen contents.

Dry weight CRs, based on lichens instead of rumen contents, dropped from 15% to 5% for U and from 320% to 145% for ²²⁶Ra, but rose from 30% to 43–56% for ²¹⁰Pb and from 12% to 41–51% for ²¹⁰Po. These differences illustrate two biases in the CR data, based on rumen contents: a) all rumen contents were below the detection limit of 20 ppb for U and thus were assigned 50% of the detection limit (10 ppb), which may overestimate the resulting

U CR; and b) ²¹⁰Po and ²¹⁰Pb CRs, based on rumen contents, are too low if the radionuclides are absorbed from the rumen during digestion.

The food chain CRs for ¹³⁷Cs were greater than unity, showing that ¹³⁷Cs bio-magnified or increased in concentration with trophic level as one moves up the food chain from rumen contents to caribou. Uptake of ¹³⁷Cs, as a K analog, is inversely proportional to the available K in the environment. Thus, organisms in environments poor in K, such as tundra and the southeastern United States, accumulate ¹³⁷Cs readily (15). Observed ratios (i.e., ¹³⁷Cs/⁴⁰K in average caribou tissue relative to the ¹³⁷Cs/⁴⁰K ratio in rumen contents) take this nutrient competition into account. Observed ratios were also over unity for the Wollaston caribou, ranging from 2.1 to 3.4; they were also above unity (range 3.2–3.6) if muscle was used in the

calculation rather than average caribou tissue. Thus, forage for Wollaston caribou was relatively poor in K, allowing for biomagnification.

Food chain CRs above unity also occurred for ²²⁶Ra. The effect of ²²⁶Ra storage in bone raised the average caribou concentration to 3.8 Bq/kg relative to rumen contents (0.83 Bq/kg). However, long-term ²²⁶Ra accumulation in bone rather than true biomagnification raised the ²²⁶Ra CR. Thus, the CR for average caribou tissue, which included bone, was 3–5 versus a CR for muscle alone of only 0.11–0.25.

To remove the biases associated with bone accumulation of radionuclides, food chain CRs were also calculated from rumen contents or lichens to caribou muscle only (Table 4). The CRs were 1–3% for U, 6–7% for ²²⁶Ra, 1–2% for ²¹⁰Pb, and 22–26% for ²¹⁰Po and showed that ²¹⁰Po is the uranium series radionuclide most

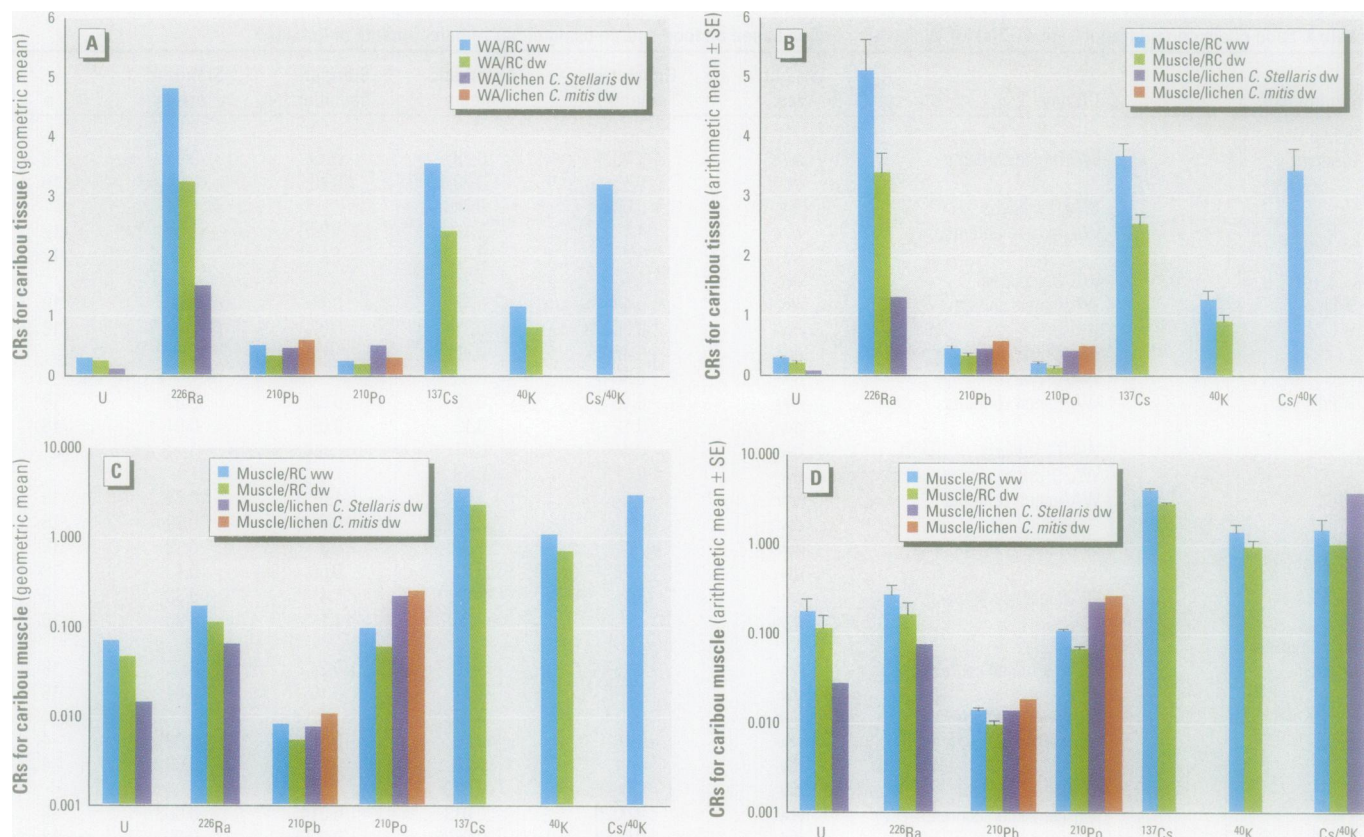


Figure 8. Food chain concentration ratios (CRs) in Wollaston caribou tissues based on wet weight versus dry weight and the type of lichen in the diet (*Cladonia stellaris* or *C. mitis*). Abbreviations: dw, dry weight; RC, rumen contents; SE, standard error; WA, weighted average; ww, wet weight. (A) average caribou tissues (geometric means). (B) Average caribou tissue (mean and SE). (C) Caribou muscle (geometric means). (D) Caribou muscle (mean and SE).

efficiently transferred to caribou meat. Muscle/lichen CRs tended to be lower with *C. stellaris* lichens because these lichens had higher radionuclide concentrations than *C. mitis* as a result of species-specific bioaccumulation and their location within the Wollaston mining area.

The CRs for caribou muscle proved to be the most convenient and least biased measures of food chain transfer because *a*) they did not require the calculation of weighted average caribou concentrations; *b*) they avoided the problem of long-term accumulation of ^{226}Ra and ^{210}Pb in bone; and *c*) they aid in estimating radionuclide transfer from lichens (which are easily monitored around uranium mines or fallout zones) to the tissues most commonly consumed by people.

Radiation doses from caribou consumption. Mean concentrations of radionuclides in Wollaston caribou meat were highest for ^{137}Cs (370 Bq/kg) followed by ^{40}K (78 Bq/kg), ^{210}Po (14 Bq/kg), ^{210}Pb (1.1 Bq/kg), ^{226}Ra (0.21 Bq/kg), and U (0.040 Bq/kg; Table 2). These concentrations (in becquerels per kilogram) were multiplied by an estimated annual intake (in kilograms per year) and a dose conversion factor (DCF in sieverts per becquerel) from the

International Commission on Radiological Protection (ICRP) (7,16,17) to yield an annual effective dose, shown in millisieverts per year in Table 6. The DCFs take into account the absorption, distribution, and physical and biological half-lives of each radionuclide in the human body.

For Wollaston Post residents consuming 100 g of caribou meat/day, ^{210}Po , followed by ^{137}Cs , contributed most of the radiological dose (0.85 mSv/year; Figure 9). This is because absorption of ^{210}Po is estimated at 50% and, once absorbed, it decays rapidly (half-life = 138 days) via the emission of alpha particles. ^{137}Cs absorption is 100%, but the longer half-life (30 years) and its beta and gamma emissions make its contribution to radiological dose less effective. The additional consumption of organ meats (i.e., one caribou liver and 10 kidneys/year) along with caribou meat doubled the radiation dose, with ^{137}Cs making less and ^{210}Pb making more of a contribution.

A 1-year-old child assumed to consume 10 g meat/day, 10% of the adult intake, received a dose of 0.48 mSv/year (i.e., more than half of the adult dose). The elevated DCF used for children reflects the greater absorption rate of ^{210}Po in the GI tract,

which doubles the dose rates per becquerel of intake in children versus adults.

The doses, calculated above, are annual doses. Because most of the dose is from short-lived ^{210}Po , the dose is delivered within 1 year of intake. Because caribou intake is chronic in northern Canada, the doses delivered over a lifetime of 70 years are essentially 70 times the annual dose.

Radiological risk assessment. A risk coefficient of one fatal cancer per 10 persons exposed to an acute dose of 1 Sv (1×10^{-1} person-Sv) has been recommended (18). This coefficient can be extrapolated down to a lower dose of 1 mSv, assuming a linear model with no threshold and a dose rate effectiveness factor of 2 (i.e., that 50% of the genetic damage caused by radiation-induced free radicals can be repaired at low doses). On the basis of these assumptions, a dose of 0.85 mSv/year for adults consuming 100 g caribou meat/day is 4.25 fatal cancers/100,000 persons consuming caribou at this rate ($0.85 \text{ mSv} \times 5 \text{ cancers/100,000 person-mSv} = 4.25 \times 10^{-5}$). This risk is doubled to 8.5×10^{-5} if 1 liver and 10 kidneys/year are also consumed because the dose is doubled from 0.85 to 1.7 mSv/year. The lifetime risk of fatal cancer is increased by a factor of 70 to 6×10^{-3} (6 cancers per

1,000) if a lifetime of caribou intake at this rate is assumed over 70 years. These risk estimates can be further raised or lowered depending on the intake rate assumed and the levels of radionuclides in edible caribou tissues found in different regions.

Discussion

The affinity of ^{210}Po for soft tissues enhances food chain transfer and radiation doses in the lichen–caribou–human food chain far more than any other radionuclide. Although muscle concentrations of ^{137}Cs in Wollaston caribou (370 Bq/kg wet weight) are relatively high compared to other Canadian caribou, ^{210}Po delivers three times the radiation dose of ^{137}Cs via caribou meat ingestion. Although ^{210}Po does not biomagnify like ^{137}Cs and DCFs from the ICRP (17) assume only 50% absorption in adults, the short half-life and greater biological effectiveness of alpha radiation are responsible for the ability of ^{210}Po to enhance the background doses in northern Canadians consuming caribou.

^{210}Po analyses of fur and pancreas were an opportunity to test the hypothesis that ^{210}Po may function as a sulfur analog in proteins, which are rich in sulfhydryl groups. For example, high ^{210}Po levels were found in goat hair (19) and insulin (20). In the Wollaston Lake caribou, ^{210}Po concentrations were higher in fur (65 Bq/kg) and pancreas (79 Bq/kg) than in blood (28 Bq/kg) or muscle (14 Bq/kg), but were lower than liver (332 Bq/kg) and kidney (156 Bq/kg). $^{210}\text{Po}/^{210}\text{Pb}$ ratios in spleen (5.3) were much higher than in fur (0.18).

^{210}Pb concentrations in bone showed little correlation with age, which was also the case with Baker Lake and Snowdrift caribou (3,4). Lack of ^{210}Pb accumulation in caribou bone with time is probably due to bone turnover and/or loss of ^{210}Pb in the annual cycle of antler growth and drop. ^{210}Pb accumulation is primarily in the surface and exchangeable compartments of bone, where it is removed during remodeling by osteoclasts, rather than in the bone volume. Salmon et al. (21) found that ^{210}Pb in the 18 Wollaston caribou bones was short-lived and resided primarily in the outer 1 μ of the bone surface. This had little impact on radiological doses to the two radiosensitive tissues associated with bone, red marrow, and bone surface epithelia because the α -emitting ^{210}Po did not have time to grow into equilibrium with the short-lived, surficially deposited ^{210}Pb .

Thomas et al. (3,4) found that caribou from the tundra region near Baker Lake had significantly more ^{210}Po and ^{210}Pb than caribou from the subarctic boreal forest near Snowdrift. This is due to the predominance

of the caribou forage lichen *Cetraria nivalis* in the Baker Lake tundra, which has a larger surface area for accumulating ^{210}Po and ^{210}Pb than the predominant forage lichen

in subarctic boreal forest (*Cladonia mitis*) found in Snowdrift and northeastern Saskatchewan. Because of these differences in forage, Wollaston caribou should have

Table 5. Radionuclide concentrations in lichens^a used for food chain concentrations ratios for Wollaston caribou.

Lichens	U (ppb)	^{226}Ra (Bq/kg)	^{210}Pb (Bq/kg)	mPo ^b (Bq/kg)
<i>C. stellaris</i> (dry weight)				
1994 Wollaston Post site	111	6.0	200	80
1994 Hungry Island site	129	5.0	300	180
1994 Parker Lake site	368	20	350	300
1994 Burned Peninsula site	1,400 ^c	10	450	250
1994 North of Torwalt Lake site	486	18	400	300
1994 Upstream of McLean Lake site	304	4.7	350	300
1994 Henday Lake site ($n = 3$)	127	14	333	450
1994 Points North Power Station site ($n = 3$)	77	11	250	200
Arithmetic mean	229	11	329	258
Standard deviation	157	6	80	109
Standard error	59	2	28	3
n	7	8	8	8
Geometric mean	186	10	320	233
<i>C. mitis</i> (dry weight)				
1991 Kasba Lake site K11			200	350
1991 Kasba Lake site K12 ($n = 2$)			275	190
1991 Kasba Lake site K13			120	70
1991 Kasba Lake site K14			250	250
1991 Kasba Lake site K21			300	250
1991 Kasba Lake site K22			300	300
1991 Kasba Lake site K23 ($n = 2$)			185	155
1991 Kasba Lake site K24			250	250
1991 Kasba Lake site K31			250	140
1991 Kasba Lake site K32			250	200
1991 Kasba Lake site K33			250	250
1991 Kasba Lake site K34			400	180
Arithmetic mean			253	215
Standard deviation			69	75
Standard error			20	22
n			12	12
Geometric mean			243	201

^aLichen species include *Cladonia stellaris* lichens from Wollaston Lake and *C. mitis* from Kasba Lake, north of Wollaston Lake. ^bMeasured ^{210}Po values not corrected for the time between sampling and counting. ^cThis outlier value was omitted from analyses.

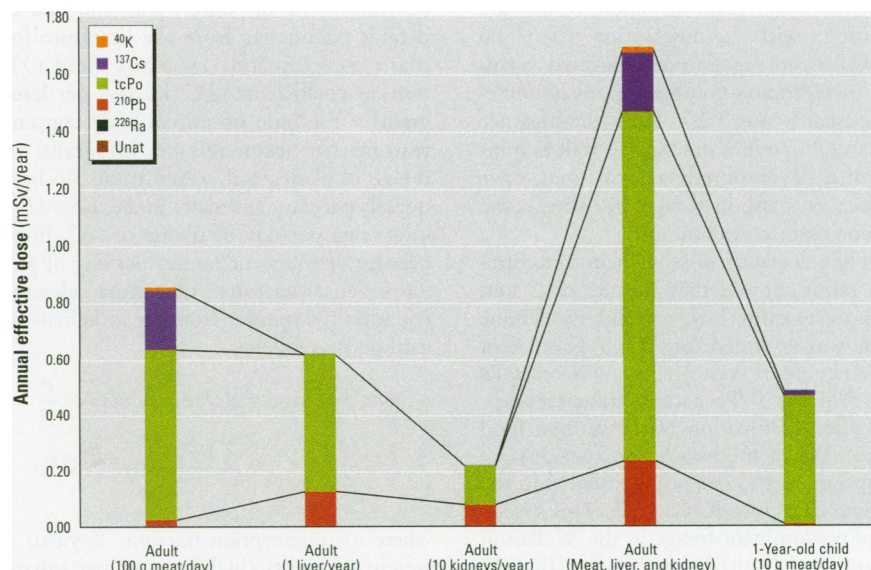


Figure 9. Annual effective doses for people consuming Wollaston caribou. Abbreviations: tcPo, time-corrected ^{210}Po concentration; Unat, natural uranium.

Table 6. Effective annual doses to people consuming Wollaston caribou meat, liver, and/or kidney.

Radionuclide	Adult			Meat, liver, and kidney	1-Year-old child (10 g meat/day)
	100 g meat/day	1 liver/year	10 kidneys/year		
Intake rate (kg/year)	36.525	1.2	0.687		3.6525
Uranium					
Bq/kg ww	0.040	0.057	0.44		0.040
DCF (Sv/Bq)	4.75×10^{-8}	4.75×10^{-8}	4.75×10^{-8}		1.25×10^{-7}
Dose (mSv/year)	6.9×10^{-6}	3.2×10^{-7}	1.4×10^{-6}	8.7×10^{-6}	1.8×10^{-5}
²²⁶ Ra					
Bq/kg ww	0.21	1.7	1.2		0.21
DCF (Sv/Bq)	2.8×10^{-7}	2.8×10^{-7}	2.8×10^{-7}		9.7×10^{-7}
Dose (mSv/year)	2.1×10^{-3}	5.7×10^{-4}	2.3×10^{-4}	2.9×10^{-3}	7.4×10^{-4}
²¹⁰ Pb					
Bq/kg ww	1.1	154	169		1.1
DCF (Sv/Bq)	7.0×10^{-7}	7.0×10^{-7}	7.0×10^{-7}		3.6×10^{-6}
Dose (mSv/year)	2.8×10^{-2}	1.3×10^{-1}	8.1×10^{-2}	2.4×10^{-1}	1.4×10^{-2}
²¹⁰ Po (time corrected)					
Bq/kg ww	14	336	156		14
DCF (Sv/Bq)	1.2×10^{-6}	1.2×10^{-6}	1.2×10^{-6}		8.8×10^{-6}
Dose (mSv/year)	6.1×10^{-1}	4.8×10^{-1}	1.3×10^{-1}	1.2	4.5×10^{-1}
¹³⁷ Cs					
Bq/kg ww	370	232	557		370
DCF (Sv/Bq)	1.4×10^{-8}	1.4×10^{-8}	1.4×10^{-8}		1.2×10^{-8}
Dose (mSv/year)	1.9×10^{-1}	3.9×10^{-3}	5.4×10^{-3}	2.0×10^{-1}	1.6×10^{-2}
⁴⁰ K					
Bq/kg ww	78	81	89		78
DCF (Sv/Bq)	6.2×10^{-9}	6.2×10^{-9}	6.2×10^{-9}		6.2×10^{-9}
Dose (mSv/year)	1.8×10^{-2}	6.0×10^{-4}	3.8×10^{-4}	1.9×10^{-2}	1.8×10^{-3}
Total dose (mSv/year)	0.85	0.62	0.22	1.68	0.48

Abbreviations: ww, wet weight; DCF, dose conversion factor. DCFs, or ingestion dose coefficients, were taken from the ICRP (17) for ²²⁶Ra, ²¹⁰Pb, ²¹⁰Po, and ¹³⁷Cs and from the ICRP (16) for ⁴⁰K; the U DCF was taken to be an average of the ²³⁴U and ²³⁸U DCFs (5.0×10^{-8} and 4.5×10^{-8} Sv/Bq) since both contribute equal activities in the natural U measured in ppb. Weight of a single Wollaston caribou kidney = 68.7 g ($n = 15$).

lower concentrations than Baker Lake caribou, and this was generally the case.

The one exception was ²¹⁰Pb in kidney, which was significantly higher in Wollaston caribou than in either Baker or Snowdrift caribou. Wollaston caribou also had lower ²¹⁰Po/²¹⁰Pb ratios in both bone and kidney than the other caribou. These results are consistent with the observation that ²¹⁰Pb in Wollaston vegetation is elevated versus that in vegetation in other regions of northern Saskatchewan (22). Thus, the presence of natural uranium outcrops as well as mining and development activity may have influenced caribou forage and thus some caribou tissue concentrations.

The calculation of food chain concentration ratios showed that transfer of U and ²²⁶Ra were quite low, particularly if bone tissue was excluded for ²²⁶Ra. The use of rumen contents versus lichens lowered CRs for ²¹⁰Pb and ²¹⁰Po because rumen absorption affects estimation of the caribou food source. When the two lichen species were compared, more credence was placed in the *C. mitis* data from Kasba Lake. This species is the predominant forage in the Wollaston region, and limited data suggest that *C. mitis* may be less efficient at accumulating airborne radionuclides than *C. stellaris* (3,4).

The use of these site-specific food chain CRs is evident if one examines the common environmental pathways models used to predict plant and animal concentrations for environmental impact assessments. One such model (23) predicts concentrations by using *a*) air deposition rates (AD); *b*) concentrations in air, water, and soil; and *c*) default parameters from the literature for plant interception (r), plant yield (Y), transfer coefficients (TC; in days per kilogram) = the ratio of animal body concentrations (in becquerels per kilogram) to intake of plants, soil, water, or air (in becquerels per day) and daily intake rates (F = kilograms per day of plants or soil, liters per day of water, or meters per day of air) via seven equations. The most relevant equation for transfer from air to lichens to caribou is as follows:

$$\text{Plant ingestion} = AD(Bq/m^2/day) \times \frac{r}{Y} \times \frac{1 - e^{-\lambda t}}{\lambda} \times TC \times F,$$

where r = interception fraction, Y (yield) = weight of plants (in kilograms per square meter), λ = fraction lost per day = $\ln 2$ /effective half-life of the radionuclide in days,

and t = length of above-ground growing season in days.

The results from such a model suffer from the lack of knowledge/measurement of site-specific air deposition rates, transfer coefficients, and animal intake rates. The first three terms of the equation can be replaced by direct measurements of lichens as caribou winter forage. The use of average caribou tissue/lichen or muscle/lichen CRs, as calculated in this study, can replace the reliance on default transfer coefficient and animal intake rates, which are often known only for other species and other locations. Thus, the routine monitoring of lichens and calculation of caribou/lichen CRs improves the accuracy and predictive power of environmental impact and health risk assessments for this important northern food chain. If ²¹⁰Pb and ²¹⁰Po concentrations in lichens doubled in the Wollaston region due to uranium mining activities, then the human doses from consumption of ²¹⁰Po with caribou could double. Should this happen, people would receive an incremental dose of 1 mSv from human activities, which would be subject to the international regulatory limits for public exposure (23).

The doses and risks, calculated in this study for people consuming Wollaston caribou, are low compared to estimates up to 14 mSv/year for more northern regions of Canada, where currently there is no active uranium mining (8). The higher doses result from the higher caribou intake rates assumed and the higher ²¹⁰Po concentrations in caribou muscle, liver, and kidney measured in these regions, particularly in Baker Lake, where natural U outcrops occur. For the most critical group, a dose of 14 mSv translates into a risk of fatal cancer of 7 per 10,000 exposed per year and 5 per 100 exposed over a 70-year lifetime. These lifetime risks from a natural source of background radiation are significant but still small as compared to the lifetime risk of cancer mortality from all sources, estimated at 1 in 5 for northern Saskatchewan aboriginal communities (24,25) or the slightly higher rate of 1 in 4 in Canada as a whole (26,27). Lifetime cancer incidence and mortality, respectively, are 41.2% and 27% in males and 35.5% and 22.5% in females in Canada (27).

The default risk coefficients used to determine the above cancer risks incorporate a factor for DNA repair mechanisms, which generally function at low doses. However, the high linear energy transfer (LET) of the alpha radiations from ²¹⁰Po cause more double-strand breaks in DNA, which are difficult to repair accurately (28,29). The alpha radiation damage from ²¹⁰Po often results in cell death. Because of cell-killing

effects, the carcinogenic potential of ^{210}Po may result more from promotion than from initiation processes (30). In addition, ^{210}Po appears to behave as a sulfur analog, which binds or replaces S in free-radical scavenger molecules such as glutathione or metallothionein. Thus, the affinity of ^{210}Po to bind to sulfhydryl groups may also inhibit repair of the damage caused by its radiological decay. All of these considerations simply point out the uncertainty in using default risk coefficients, which may be more appropriate for the damage caused by low LET radiations from beta- and gamma-emitting radionuclides such as ^{137}Cs versus the alpha emitters such as ^{210}Po .

Conclusions

Radionuclide concentrations of U, ^{226}Ra , ^{210}Pb , ^{210}Po , ^{137}Cs , and ^{40}K were measured in tissues from 18 Wollaston caribou. Uranium was near detection limits, being above detection in all liver and fecal samples and three to four samples each of bone, kidney, and muscle. Most of the ^{226}Ra body burden was in bone. ^{210}Pb was primarily in bone, followed by kidney and liver, with kidney levels significantly enhanced as compared to previous measurements in other Canadian caribou. ^{210}Po exceeded concentrations of its precursor, ^{210}Pb , in all soft tissues. Because ^{210}Po , ^{137}Cs , and ^{40}K were present in edible soft tissues, human consumption of these tissues enhances the transfer of these radionuclides through the food chain.

Food chain transfer was determined as percentages using CRs, where CR = concentrations in average caribou tissue or muscle divided by the concentration in rumen contents and previously collected lichen samples as the caribou food source. Transfer from dry lichens to dry caribou muscle was 1–3% for U, 6–7% for ^{226}Ra , 1–2% for ^{210}Pb , and 22–26% for ^{210}Po ; the transfer from rumen contents to caribou muscle was 5–11% for U, 11–16% for ^{226}Ra , 0.5–1.3% for ^{210}Pb , 6% for ^{210}Po , and 76–90% for ^{40}K , but 250–260% for ^{137}Cs . Observed ratios of $^{137}\text{Cs}/^{40}\text{K}$ in muscle to rumen contents showed that ^{137}Cs biomagnifies by a factor of 3.2–3.6.

These CRs provide new information on U, ^{226}Ra , ^{210}Pb , and ^{210}Po transfer at a crucial site in northern Saskatchewan for the evaluation of uranium mining impacts in a critical food chain. Dry weight muscle/lichen CRs were the most free of bias and the most useful because they a) eliminated the effect of moisture and prior absorption of radionuclides in rumen contents; b) eliminated the effect of long-term

bone accumulation of ^{226}Ra and ^{210}Pb when a weighted average of caribou tissues was calculated; and c) allowed easy prediction of caribou meat concentrations from lichen concentrations for environmental pathways modeling and human radiological risk assessment. Lichen levels of ^{210}Pb and ^{210}Po would have to double in the Wollaston area of northern Saskatchewan before people consuming caribou would receive a dose increment of 1 mSv/year as a result of uranium mining and for the industry to require regulation.

Annual effective doses from the consumption of 100 g/day of caribou meat by northern Canadian adults were 0.85 mSv/year. Additional consumption of 1 liver and 10 kidneys per year doubles that dose to 1.7 mSv/year. A 1-year-old child, consuming only 10% of the adult intake of caribou meat, receives more than half the adult dose due to greater absorption of ^{210}Po . These doses are predominantly from ^{210}Po , which far exceeds the fission product ^{137}Cs as the main contributor to natural background radiation dose from consuming caribou in northern Canada.

The risk of fatal cancer from a dose of 1.7 mSv is 8.5×10^{-5} per year, and 6×10^{-3} over a 70-year lifetime if caribou meat, liver, and kidney are consumed at the rate postulated. Doses and risk are almost 10 times higher in other regions of Canada where ^{210}Po concentrations in caribou are higher and if higher intake rates are assumed.

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